

March 2014

Science & Technology

REVIEW



Innovations in Radiochemistry

Also in this issue:

Light, Compact Neutron Detection

Biosecurity in a Chip

Improving Nuclear Fuel Burnup

About the Cover

Over the years, radiochemistry has proven to be a vital field for research innovation in support of Livermore's national security mission. In past work for the Laboratory's nuclear weapons program, radiochemists developed methods to capture and analyze radioactive debris produced in underground experiments, allowing weapons designers to better understand the physics involved in the detonation process. Today, radiochemistry research at Livermore has grown to encompass a wider range of national security and scientific missions, such as nuclear forensics and nuclear reactor safety. The article beginning on p. 4 describes many of the recent accomplishments. On the cover, nuclear chemist Narek Gharibyan prepares to analyze a sample that was produced at the Center for Accelerator Mass Spectrometry.



Cover design: George A. Kitrinos

About S&TR

At Lawrence Livermore National Laboratory, we focus on science and technology research to ensure our nation's security. We also apply that expertise to solve other important national problems in energy, bioscience, and the environment. *Science & Technology Review* is published eight times a year to communicate, to a broad audience, the Laboratory's scientific and technological accomplishments in fulfilling its primary missions. The publication's goal is to help readers understand these accomplishments and appreciate their value to the individual citizen, the nation, and the world.

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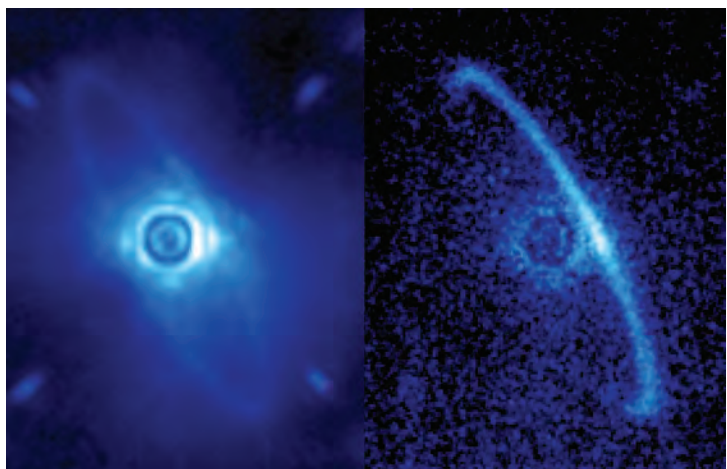
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First Light for Gemini Planet Imager

In November 2013, after nearly a decade of work by an international collaboration, the Gemini Planet Imager (GPI) began collecting light from planets outside our solar system. GPI (pronounced gee-pie) is the first astronomical camera designed to directly detect light from extrasolar planets (exoplanets) or dusty disks that are 1 million to 10 million times fainter than the stars they orbit. The GPI team, which includes researchers from Lawrence Livermore, released the first images at the January 2014 meeting of the American Astronomical Society.

“These early first-light images are almost a factor of 10 better than the previous generation of instruments,” says Livermore astrophysicist Bruce Macintosh, who led the collaboration under the direction of the Gemini Observatory. “In one minute, we were seeing planets that used to take us an hour to detect.”



About the size of a small car, GPI is deployed on the 8-meter-diameter Gemini South telescope near the summit of Cerro Pachón in Chile. The imager’s sophisticated adaptive optics (AO) system is designed to eliminate the blurring effects caused by turbulence in Earth’s atmosphere. The AO system has a 2-centimeter-square deformable mirror with 4,000 actuators that correct for atmospheric distortions by adjusting the mirror’s shape 1,000 times per second with an accuracy of better than 1 nanometer. Manufacturing the mirror with etched silicon instead of reflective glass reduced its size and improved its stability.

“GPI’s performance requirements are extremely challenging,” says Livermore engineer Lisa Poyneer, who developed algorithms and led testing for the AO system. “As a result, the system features several original technologies that were designed specifically for exoplanet science.”

For the first-light observations, the GPI team targeted previously known planetary systems, including the four-planet HR8799 system and Beta Pictoris. The researchers also operated

the imager in polarization mode, which is tuned to look at starlight scattered by tiny particles, allowing them to study a ring of dust orbiting the very young star HR4796 (shown in the image below left). Previous instruments could detect only the edges of this dust ring, which may be the debris remaining from planet formation, but GPI can follow the ring’s entire circumference.

Imaging exoplanets complements other astronomical projects, such as NASA’s Kepler Mission. Former Livermore postdoctoral researcher Dmitry Savransky, who worked on GPI before moving to a position at Cornell University, notes, “Broad survey missions such as Kepler have revealed the variety of planets that exist in our galaxy. GPI will allow us to study a few dozen planets in exquisite detail.”

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Study Shows an Explosive Responding to Shock Waves

A team of Livermore researchers has combined ultrafast time-resolved experiments with molecular dynamics simulations and thermochemical calculations to reveal how an explosive responds to a high-pressure shock from the moment of impact to the time of detonation. The results, which are featured on the cover of the December 12, 2013, issue of the *Journal of Physical Chemistry A*, represent a milestone in understanding chemical initiation and detonation.

The team worked with hydrogen peroxide, which is composed of one oxygen–oxygen bond and two oxygen–hydrogen bonds within a hydrogen-bonding network. Sorin Bastea, who led the research team, says, “Hydrogen peroxide is a relatively simple molecular liquid that gives us the opportunity to study a very complex process.” Livermore physical chemist Nir Goldman, who along with Will Kuo led the project’s simulation efforts, adds, “What is unique about this research is that we have experimental data that corroborate our theoretical predictions on the exact same timescale.”

In shock-wave experiments led by Mike Armstrong and Joe Zaug, a 0.001-millimeter-thick aluminum film in contact with the peroxide was hit with a very short burst of laser energy. (See the video at www.llnl.gov/news/newsreleases/2013/Dec/videos/1202_detonation.mpg.) Using optical interferometry, the researchers then measured the speed of the shock wave as it traveled through the fluid. The peroxide began to tear apart 50 picoseconds (50-trillionths of a second) after the sample was shocked, and by 100 picoseconds, chemical bonds were completely broken. The temperature increased by more than 1,500 degrees, and the explosive pressure wave spiked to more than 200,000 atmospheres (20 gigapascals). Thermochemical calculations showed that the amount of chemical reaction in the experiments was approximately 50 percent.

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Reinvigorating a Signature Asset

WITHIN the nuclear weapons design laboratories such as Lawrence Livermore and Los Alamos, the historic mission for nuclear and radiochemists was to support the nuclear test program. Our chemists developed and implemented techniques to gauge the performance of nuclear devices by analyzing the radioisotopes produced. This work led to advances in many areas of radiochemistry, such as chemical separation science, radiation detection, and instrument automation. Nuclear testing may have ended two decades ago, but nuclear and radiochemistry remain among the Laboratory's signature capabilities. The opportunities for applying radiochemistry expertise to pressing national needs have, if anything, expanded, especially over the past few years. Ongoing research performed by our outstanding nuclear and radiochemists has applications in national and global security, energy security, environmental stewardship, and human health.

My involvement with the Laboratory's radiochemistry experts and resources began during my tenure as director of Livermore's Forensic Science Center. In the aftermath of September 11, 2001, agencies in Washington, DC, including the newly established Department of Homeland Security, showed an increased interest in forensic science and the unique capabilities at Livermore and the other national laboratories. Livermore worked closely with these organizations to determine how the Laboratory's expertise might be applied to chemical, biological, and nuclear forensic analysis.

As described in the book *Nuclear Forensic Analysis*, by Livermore scientists Ken Moody, Ian Hutcheon, and Pat Grant, "The primary technical objective of nuclear forensic analysis is to determine the attributes of questioned radioactive samples. Reduced to simple terms, the salient forensic questions for a nuclear sample are: What is it? What was its origin? How did it get there? Who was involved?" Answering these questions requires not only a fundamental understanding of radiochemistry and nuclear science but also experience with real-world materials and samples, which makes nuclear forensics a natural fit with the Laboratory's strengths.

With surging interest in radiochemistry research, Livermore's management found that the infrastructure supporting this work needed modernizing to better serve the diverse customer base. Building 151, the home of Livermore radiochemistry, houses a one-of-a-kind nuclear counting facility and laboratories designed

specifically for radiochemistry experiments, but these facilities were built in another era to support one programmatic mission. Meeting the multi-programmatic national nuclear security challenges of the 21st century has led us to improve or build new laboratories in the facility. The improvements thus far have paid dividends, and we are planning to modernize other laboratories as funding is identified. Already, the revitalized program has allowed us to better support existing projects and sponsors and has attracted new researchers, collaborators, and sponsors.

In addition to reinvesting in our radiochemistry capabilities, we have been examining how we can transcend traditional disciplinary and organizational boundaries to better leverage other Laboratory capabilities, including mass spectrometry, environmental radiochemistry, and diagnostic development. For instance, we have fostered closer collaboration between staff at the Center for Accelerator Mass Spectrometry—an invaluable research tool—and radiochemistry researchers. Responding flexibly to changing national priorities while delivering the outstanding science and engineering results expected of us requires strategic investment in our infrastructure, associated capabilities, and staff.

A reinvigorated radiochemistry program and facilities benefit the entire Laboratory and beyond. As described in the article beginning on p. 4, Livermore scientists are pursuing a number of innovative nuclear and radiochemistry research efforts with wide-ranging application. For instance, new techniques for characterizing radioactive target components are generating high-fidelity data from stockpile stewardship experiments, and careful measurements of nuclear reaction products may eventually benefit nuclear reactor safety. An automated chemical separation technique will enhance our capabilities for experimental diagnostics at the National Ignition Facility, nuclear forensics in support of national security, and the search for superheavy elements. Finally, by implementing more realistic nuclear forensics exercises, we will be better able to rapidly and accurately identify nuclear materials.

Radiochemistry is again stepping into the forefront of science at the Laboratory. Livermore's nuclear and radiochemists will undoubtedly make key contributions to the institution's missions in the years ahead.

■ Glenn A. Fox is associate director for Physical and Life Sciences.



Radiochemistry Renaissance

Radiochemists apply nuclear test experience and unique capabilities to new national security and scientific challenges.

ABOUT a century ago, the enduring dream of alchemists was realized when physicist Ernest Rutherford and his collaborators demonstrated that one element can change into other elements through the process of radioactive decay. Rather than turning lead into gold, Rutherford's discovery led to the founding of radiochemistry—the study of radioactive isotopes—and nuclear chemistry, which focuses on the properties of atomic nuclei and the processes involved in element transformation. Together, these fields form a pillar that supports Lawrence Livermore's nuclear security mission, a role they have held since the institution was founded in 1952. (See *S&TR*, June 2002, pp. 24–30.)

Radiochemists provided crucial contributions to the Laboratory's nuclear weapons test program. They created tracer components for test devices by using actinides—radioactive elements with atomic numbers from 89 through 103, the most common of which is uranium. They also assessed a weapon design's performance by studying the radioactive debris and gases produced in an experiment. Since the moratorium on underground nuclear testing began in 1992, radiochemistry research at Livermore has grown to encompass a wider range of national security and scientific missions.



Livermore scientists have engaged in radiochemistry, nuclear chemistry, and heavy-element research since the Laboratory's founding in 1952. In the photo above, Wes Hayes works in hot cells on debris collected from the Hutch Event, a 1969 underground nuclear experiment that produced large quantities of rare, heavy isotopes for debris analysis. At left, Roger Henderson prepares a uranium sample for electrodeposition onto a stub for testing a resonance ionization mass spectrometry technique.

Nuclear chemist Dawn Shaughnessy, who leads the experimental and nuclear radiochemistry group in the Physical and Life Sciences Directorate, notes that many potential collaborators and project sponsors are surprised by the breadth of radiochemistry capabilities at the Laboratory. Says Shaughnessy, "In the past, Livermore radiochemistry to most people really meant nuclear test analysis, although even in the heyday of underground testing, that was only part of the endeavor."

Today, Lawrence Livermore offers a unique recipe for radiochemistry research, combining nuclear testing expertise and resources, such as rare radioisotopes collected from past underground experiments, with robust materials handling capabilities and an array of

research and isotope production facilities, including the Center for Accelerator Mass Spectrometry (CAMS) and the National Ignition Facility (NIF). Nuclear chemist Narek Gharibyan observes, "We have places to safely perform irradiation, chemical separation, and nuclear counting, all on-site. CAMS also produces radioactive isotopes for a variety of chemistry experiments. It's unusual to have all of these facilities in one place."

Shaughnessy adds, "As radiochemists, we see NIF as a giant neutron source, one of the brightest in the world. For instance, it can help us make samples for nuclear forensics experiments." But for everything Livermore offers, the recipe would be incomplete without Shaughnessy's team of a dozen or so innovative chemists, working in such diverse areas as fission, nuclear

forensics, radioactive target fabrication, transactinide chemistry, and rapid radiochemical separations.

Mapping Fission's Outcomes

During fission, an excited nucleus splits into two (or occasionally more) lighter fragments and in the process releases by-products such as neutrons or photons in the form of gamma rays. Although the results of an individual fission event are random, the distribution pattern for the particles emitted for a given parent isotope and initiating energy is statistically predictable. Precisely determining these patterns benefits important national security efforts, from nuclear forensics investigations of postexplosion debris to nuclear reactor safety.

Gharibyan and his colleagues are refining techniques for measuring the products emitted during proton- and neutron-induced fission of several isotopes, including uranium-238. Researchers have extensively studied neutron-induced fission of uranium-238 because of its importance to nuclear power, but the proton-induced reactions are not as well understood. To examine these events, researchers stack uranium, aluminum, and yttrium foils at the end of an accelerator chamber at CAMS and bombard the target with protons accelerated to energies between

10 and 15 megaelectronvolts. The foils are thin enough for protons to pass through without losing much energy, yet thick enough to generate an adequate number of fission reactions for study. Yttrium foils act as a proton flux monitor, allowing the team to calculate the likelihood of a fission reaction occurring in the uranium foils.

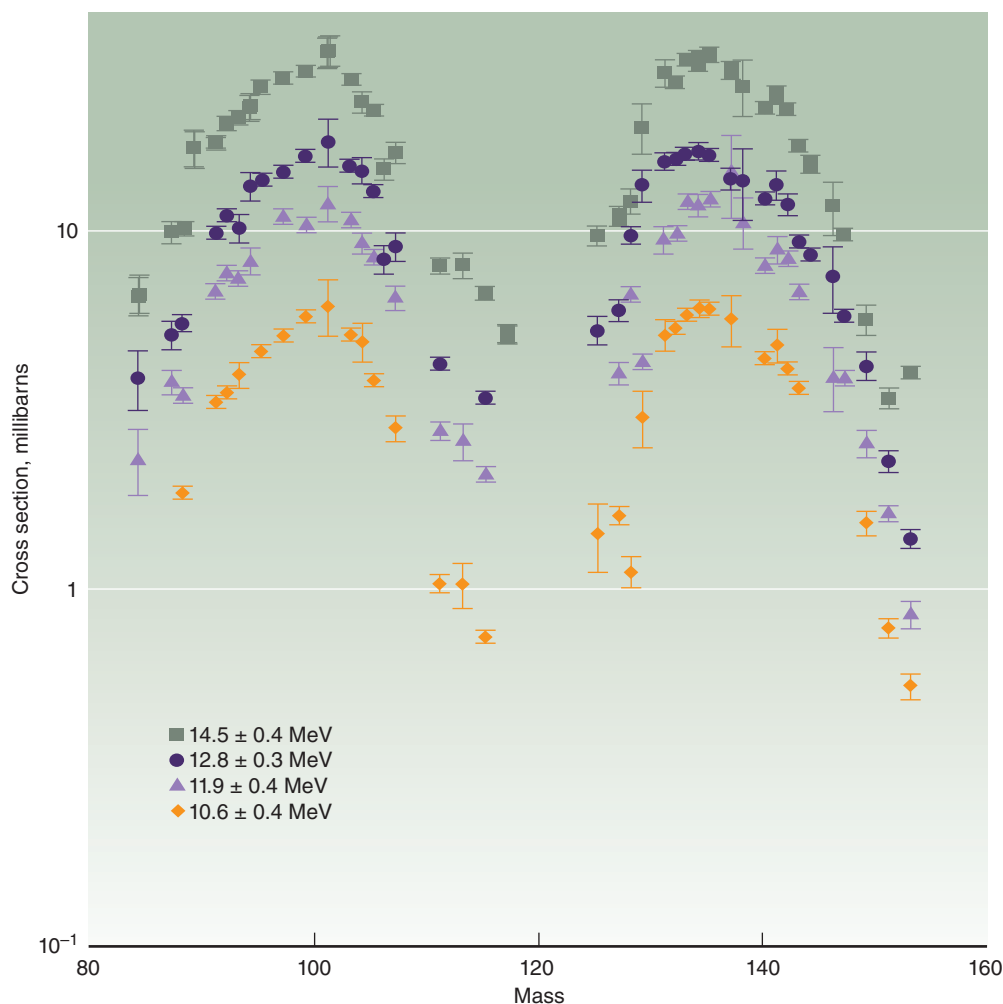
Most fission products are unstable and will decay by emission of a beta particle and characteristic gamma rays. High-purity germanium radiation detectors record the signatures of gamma rays emitted after decay, and automated analysis software processes the data to determine the identity and quantity of the gamma-emitting nuclides in the source. According to

Gharibyan, 40 to 50 spectra are collected and analyzed for each sample.

The more quickly samples arrive at Livermore's nuclear counting facility, the more radioactive products researchers are likely to detect. For many of the team's early experiments, samples received only 10 minutes of proton irradiation, to limit radiation levels and obviate the need for a cooldown period before sample removal and analysis. The fast turnaround enabled researchers to detect products with half-lives as short as 5 minutes.

Longer-lived and lower-yield products experience fewer decay events and thus can be more difficult to detect. To create these elusive particles in greater

A team of nuclear chemists at Livermore is refining techniques for precisely determining the unique patterns of isobars—nuclides of different elements with the same mass number—produced by the fissioning of various parent isotopes at different energies. This graph shows the likelihood of various isobars being produced during fission of uranium-238 when the proton inducing the reaction is moving at different speeds. Colored symbols indicate proton energy measured in megaelectronvolts (MeV); 1 millibarn is equivalent to 10^{-27} square centimeters.



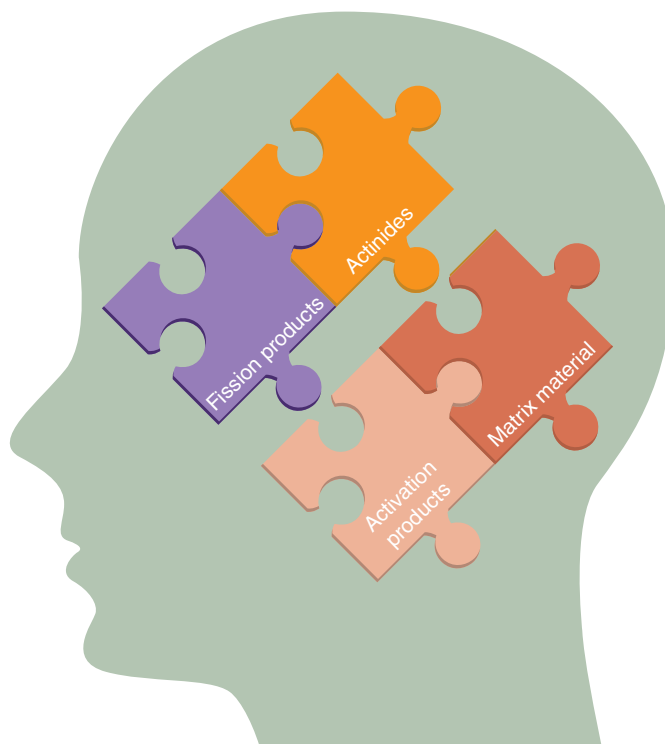
numbers, the Livermore team increased the irradiation period to a full day and the nuclear counting period to two months. After days or weeks, the interfering signatures produced by many of the shorter-lived fission products are eliminated, so that longer-lived and lower-yield products are somewhat easier to distinguish.

Chemically isolating and removing selected fission products from some samples before nuclear counting also helps the researchers fill in data gaps. “Radiochemistry allows us to detect emissions produced in small quantities or those that have interferences,” says Gharibyan. In one uranium-238 experiment, the researchers demonstrated that, by chemically separating samples, they could measure fission yields for about 50 percent more isobars—nuclides of different elements with the same mass number—than they could identify by nuclear detection alone. In future experiments, the team will measure fission product distributions for other actinide isotopes.

Mixing a Radionuclide Cocktail

If an illicit nuclear explosive were detonated on U.S. soil, nuclear forensics experts would be tasked with examining rubble from the explosion for traces of fissile material, fission products, and activation products—nearby debris or structural components of the bomb made radioactive by neutron activation. The more information experts could glean by characterizing these materials, the better able they would be to trace the weapon’s design and origin.

To ensure readiness in the event of such a scenario, research teams at Los Alamos, Pacific Northwest, and Lawrence Livermore national laboratories have, for the past decade, engaged in isotope identification exercises using simulated debris samples. Exercises last from days to weeks and involve both chemical analysis and computer modeling. Samples



Nuclear forensics readiness exercises use samples of synthetic nuclear debris to test participants’ skills at isotope identification.

The current method for preparing samples is to irradiate a single fuel in a reactor and collect the fuel and its fission products. A truly realistic sample would include all the pieces of the puzzle—fuels plus fission and activation products mixed in the right proportions and embedded in a matrix of dirt and debris.

are prepared at one facility, typically by irradiating uranium in a reactor and splitting it into chunks. The samples are then sent to the participating laboratories for analysis.

About three years ago, Livermore nuclear chemists, led by Kevin Roberts, began designing more realistic debris to further challenge their fellow chemists and modelers. A fully realistic sample would include a carefully curated selection of fuels, fission products, and activation products embedded in a matrix of dirt and debris. Drawing on the Laboratory’s experience in analyzing nuclear test debris (see *S&TR*, April/May 2012, pp. 11–13) as well as the results of Gharibyan’s fission investigations and other research efforts, the team is determining how best to create a “radionuclide cocktail” with ingredients in the right amounts and proportions.

Using a mix of different fuels and reaction energies will likely achieve the most realistic results. Laboratory researchers have access to isotopes from past experiments and can generate new materials using accelerators, test reactors, and other sources. In conjunction with the Crocker Nuclear Laboratory at the University of California at Davis, the Livermore team completed several practice runs to demonstrate sample production

in 2012 and two round-robin exercises with Los Alamos, Pacific Northwest, and the United Kingdom’s Atomic Weapons Establishment in 2013.

Nearby facilities such as CAMS and the cyclotron at Davis (about 135 kilometers or 85 miles north of Livermore) are preferred for sample preparation because many of the fission and activation products are short-lived. Samples must be created and shipped promptly so that exercise participants can measure the more ephemeral products. In fact, logistics and handling are no small part of the research project. Roberts notes that at first, his group adopted radiochemical methods from the underground test program because those approaches were established and effective. Now, the researchers want to modernize the techniques so they can prepare testing materials more efficiently and safely. “Making this kind of sample is not a trivial exercise,” says Roberts. “The debris can end up being fairly radioactive, and not many facilities can handle such materials. We can, and we couple it with the weapons knowledge here.”

As part of the sample preparation effort, Livermore researchers have partnered with scientists at the University of Nevada at Las Vegas (UNLV) to fabricate a potential matrix material made of equal parts

glass and concrete dust. The substance is then combined with fuels irradiated at a test facility at the Nevada National Security Site, and the resulting samples are characterized. The Laboratory team also is investigating the feasibility of using additive manufacturing to create samples that can be precisely reproduced, which would aid in data intercomparison.

Kimberly Budil, who manages the Nuclear Counterterrorism Program in Livermore's Global Security Principal Directorate, stresses the importance of using realistic debris samples for the readiness exercises. "It's important to challenge the participants in a way that mimics the response to an actual incident and at the same time to thoroughly test their diagnostic capabilities."

Roberts notes that his team is 5 to 10 years from routine production of truly realistic samples, but those developed for the round-robin exercises in 2015 should have materials in the proper proportions. By engaging in readiness exercises and sample development, Livermore researchers are helping to establish a significant national capability and fostering stronger partnerships with forensics specialists at other laboratories and with sponsors in the Departments of Defense and Energy.

A Source for Radioactive Targets

In addition to their work on nuclear fission and forensics, the radiochemists in Shaughnessy's group are fabricating

radioactive sources and targets for Laboratory experiments that require a broad range of isotopes. Some of these materials are created in facilities such as CAMS. Others are acquired by dipping into the Laboratory's archive of isotopes collected from various sources, including Oak Ridge and Savannah River national laboratories. The archive is one of a kind, offering such items as the world's only supply of a metastable form of americium-242 and the only separated batch of plutonium-244. Nuclear chemist Roger Henderson, who leads the fabrication efforts, says, "At Livermore, we have access to unique, irreplaceable source materials. Part of our job is stewardship of these materials because no one knows when they will be produced in experimentally usable quantities again."

For one Laboratory project, Henderson created 60 monitor foils of uranium-235 enclosed in aluminum holders to calibrate dosimeters being designed for field deployment following a nuclear accident. He also crafts targets for experiments to study nuclear reactions, most of which are relevant to both stockpile stewardship and fundamental science research. "Target making is a specialty process," says Henderson. "No two clients have identical specifications. There's always some difference in the substrate, size, amount of material, or other specifications."

Henderson often prepares targets for Livermore physicist Ching-Yen Wu, who studies neutron-induced reactions

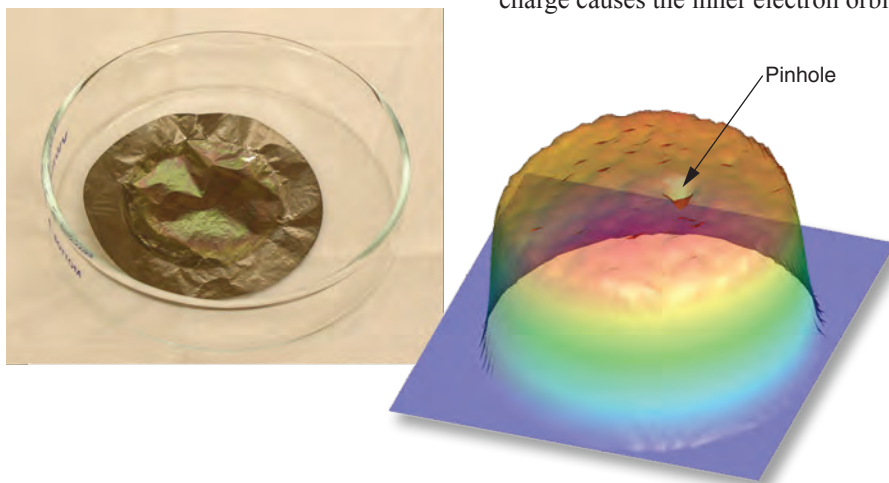
in uranium and plutonium isotopes in experiments at the Los Alamos Neutron Science Center. Wu's targets are made of freestanding foils, each only a few micrometers thick but up to 4 centimeters in diameter—quite large for heavy-element experiments. In addition, the fragile foils often require uranium or plutonium on both sides instead of the standard one side. To limit handling, Henderson developed an apparatus that electroplates both sides of the foils simultaneously. He also built an instrument that generates a three-dimensional map of the target surface, allowing researchers to characterize how smoothly and evenly the radioactive material is deposited on each surface.

Having an in-house capability to supply quality-tested radioactive sources and targets has been a boon for Livermore researchers. In particular, the electroplating technique and the surface-mapping tool have increased the consistency of radiochemistry experiments. Collaborators at GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany, are also interested in using the automated mapping system to characterize their targets.

Extracting Elemental Behavior

Other radiochemists at Livermore are using Henderson's target foils to explore the chemical behavior and fundamental properties of elements at the far end of the periodic table, which have short life spans and low production yields. Examining the transactinides, elements 104 and above, is especially challenging. Their high nuclear charge causes the inner electron orbitals

An automated surface mapping system developed at Livermore examines targets such as the foil on the left and produces a three-dimensional surface map such as the one on the right. This information allows researchers to evaluate coating consistency, which could affect an experiment, and to characterize features such as the pinhole in the foil.

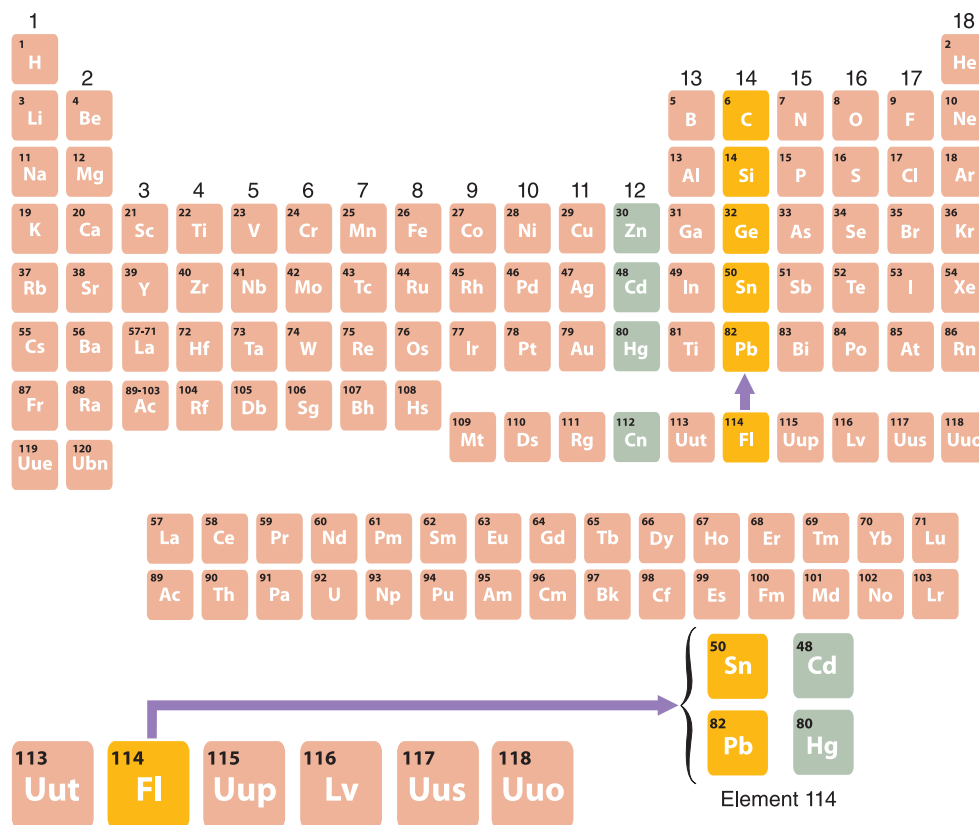


to move very close to the speed of light, which can affect an element's atomic and chemical properties. This relativistic effect may become more extreme for the heavier transactinides, making their characteristics and behavior more difficult to predict.

Six of the newest and heaviest transactinide elements were discovered by Livermore researchers in collaboration with colleagues at the Flerov Laboratory of Nuclear Reactions (FLNR) in Russia. (See the box on p. 10.) While an element's atomic number is determined by the number of protons in its nucleus, the element's placement in the periodic table is based on the chemical properties it shares with elements nearby. The question that intrigues Lawrence scholar John Despotopulos is whether the newest elements belong where they are currently situated. "Flerovium, element 114, is just beneath lead on the periodic table, so these two elements should have similar chemical properties," says Despotopulos. "But some scientists predict that flerovium is more like mercury or an inert gas such as radon."

Two gas-phase experiments of flerovium produced contradictory results. Wet-chemistry experiments, which are more complicated than gas-phase studies, would provide more insight into where the element fits in the periodic table. Despotopulos and colleagues at Livermore and UNLV are thus devising efficient and precise wet-chemistry methods to isolate the two elements. To evaluate these methods, they are using surrogates—lighter elements with putatively similar properties to the transactinides and more atoms available for study. (See *S&TR*, September 2011, pp. 4–10.)

A short nuclear lifetime and low production rate prevent flerovium from reaching chemical equilibrium in its environment. As a result, measuring the element's chemical reactivity is difficult. The Livermore and UNLV collaborators are working with carrier-free isotopes to more accurately replicate that behavior in the surrogates. In a carrier-free isotope, all the atoms of a given element consist of the same radioisotope; no stable



An element's position in the periodic table is based on its properties. Because the heaviest elements are short-lived and produced only in tiny quantities, scientists must extrapolate their characteristics by systematically studying related but longer-lived elements. For example, the properties of flerovium (Fl) may be similar to tin (Sn) and lead (Pb) or cadmium (Cd) and mercury (Hg).

isotopes are present. Carrier-free isotopes of bismuth and lead are isolated from samples of uranium-232, while antimony and tin are created at CAMS. The flerovium surrogates, lead and tin, are then mixed together, as are the element 115 surrogates, bismuth and antimony.

The research team then uses extraction chromatography to separate the desired isotopes. For this technique, a resin coated with an extractant is affixed inside a glass column along with a small amount of the sample. When the column is flushed with a solvent, usually hydrochloric or nitric acid, the component of interest adheres to the extractant. Everything else is washed through the column. The extractants selected for the Livermore experiments are crown ethers, cyclical molecules that selectively bind with certain metals.

Tests on the four surrogates in varying concentrations of solvent showed that

crown ethers extracted the targeted surrogate efficiently and precisely. Despotopulos and his colleagues are extending the extraction study to include mercury and cadmium, which may share properties with flerovium. They are also synthesizing various crown ethers to determine the best combination of extractant properties for different elements.

Chemistry at the Push of a Button

Producing enough atoms to study the heaviest transactinides requires weeks or months of accelerator beam time. Because these rare, short-lived elements can form at any time, scientists must constantly monitor the experiments. With funding from Livermore's Laboratory Directed Research and Development Program, radiochemists in Shaughnessy's group are collaborating with researchers at UNLV and Texas A&M University to design

A Smashing Collaboration

In 1989, nuclear chemists Ken Hulet from Lawrence Livermore and Georgy Flerov, founder of the Flerov Laboratory of Nuclear Reactions (FLNR) in Dubna, Russia, met at a conference and decided to work together to synthesize new elements. (See *S&TR*, October/November 2010, pp. 16–19.) Transcending international politics and intense competition within the heavy-element research community, scientists from the two institutions have built an enduring research partnership and discovered six new elements, 113 through 118.

The names chosen for elements 114 and 116, the two newest additions to the periodic table, pay tribute to that collaboration. Flerovium, the name for element 114, honors Flerov and FLNR, and livermorium, for element 116, recognizes Lawrence Livermore and its hometown, Livermore, California. Bill Goldstein, the Laboratory's deputy director for Science and Technology, says, "These names honor not only the individual contributions of scientists from the two laboratories to the fields of nuclear science, heavy-element research, and superheavy-element research, but also the phenomenal cooperation and collaboration that has occurred between scientists in the two countries."

Element discovery is a lengthy process. Before an element can be added to the periodic table and named, its existence must be detected independently at more than one laboratory and preferably through more than one method. Elements 114 and 116 were first glimpsed by the Livermore–FLNR team in 1998 and 2001, respectively. (See *S&TR*, January/February 2002, pp. 16–23.) In May 2012, the International Union of Pure and Applied Chemistry (IUPAC) officially accepted the element names. In its decision, IUPAC stated that it found the name livermorium appropriate because of Livermore's breadth of nuclear and radiochemical research.

On June 24, 2013, Laboratory employees and the city of Livermore celebrated the discovery and naming of livermorium and flerovium with a daylong celebration, attended by members of the U.S. and Russian discovery

team and Dubna city officials. The festivities began with a Laboratory-hosted colloquium, titled "Elemental Science: Livermorium and the Periodic Table." In one lecture, Livermore nuclear chemist Ken Moody discussed the history of heavy-element research and the long-standing collaboration between Lawrence Livermore and FLNR. "A collection of people with good will and shared vision can make wonderful things happen," he observed. Following the colloquium, the city of Livermore hosted a dedication ceremony at the newly renamed Livermorium Plaza and unveiled a plaque dedicated to the discovery team.

All elements with more protons than uranium, element 92, are radioactive and were discovered in a laboratory. None of these elements has a half-life long enough to have survived since Earth's formation. Essentially all transuranic elements were first synthesized at one of three facilities—FLNR, Lawrence Berkeley National Laboratory, or the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany—where inside an accelerator, a stream of lighter atoms was smashed into a heavier atom for hours or days until, finally, a pair combined. Synthesis experiments for many of the newest and heaviest elements yield only a single atom with a lifetime ranging from microseconds to a few seconds, making detection and study a challenge. Despite the difficulties, scientists persist with these investigations because each addition to the periodic table expands their knowledge of atomic structure and behavior.



A mural at Livermorium Plaza in downtown Livermore, California, commemorates the U.S.–Russian collaboration that led to the discovery of six heavy elements.

a system that captures, prepares, and measures samples with limited interaction from researchers.

The system, which is being tested at CAMS, will connect directly to the beamline where the element is produced. After screening out the incomplete fission products, it will collect complete products, dissolve them in solution, and subject them to automated chemical processing, including ion exchange or extraction

chromatography. The development team estimates that the system will reduce the time spent on chemical separation tasks by a factor of up to 10.

A system that automates some of the repetitive and time-consuming activities in chemistry has numerous other applications. Roberts, for instance, wants to apply the technique to expedite nuclear forensics research. "Urgency was not always a pressing concern for

scientists working on underground nuclear experiments," he says. "They did need to capture the short-lived fission products, but they had more time to analyze their results. We would face a much different scenario in the event of a nuclear explosion. We'd need results immediately." An automated wet-chemistry system could also be used for medical imaging and treatment, astrophysical research, or even



Livermore nuclear chemist John Despotopoulos works with the Laboratory's automated ion chromatography fraction collection system, which can chemically separate up to 12 samples simultaneously.

environmental remediation—for example, by accelerating the extraction of lead and mercury, two heavy-metal pollutants. (See *S&TR*, November 1999, pp. 17–19.)

Portability is a key design consideration for the automated system because it will likely be transported to partner facilities for element synthesis and characterization efforts. A reasonably compact system could also facilitate field deployment for nuclear forensics and environmental purposes. In a complementary effort, the team is evaluating a slimmed-down design for an extra-sensitive germanium detector to rapidly identify specific isotopes at remote locations. With improved nuclear counting techniques, the detector could identify certain isotopes in the presence of others, thus eliminating the chemical separation step—a process Roberts compares to “finding the needle without removing the haystack.”

Radiant Prospects

Budil adds that the expertise developed by Shaughnessy's group is broadening the scope of nuclear forensics research at a national level. “In the nuclear forensics community, we are continually looking

for new approaches to address evolving threats,” says Budil. “NIF is a great example. It offers unique capabilities for exploring nuclear science and nuclear chemistry at a fundamental level, but forensic scientists need to learn to apply these capabilities to our mission space. The radiochemistry team is helping us define the role for NIF experiments in current and future research.”

An effort to build a new accelerator center at Livermore has begun to gather momentum. The proposed center would include a medical cyclotron, a larger accelerator that could be used for nuclear science and as a rare isotope source, and a facility with modular laboratory spaces. “Our current facilities for nuclear science are not flexible enough for 21st-century experiments,” says Budil. “We need to be able to modify work areas to accommodate new technologies as they become available. We also want to design spaces that foster collaboration and cross-talk among scientists—not just with our teams at Livermore but with the nuclear forensics community at large.”

If the accelerator project reaches fruition, Lawrence Livermore will likely

serve as a magnet for nuclear research. In the meantime, radiochemistry researchers are working to secure beam time on other large accelerators, such as the 88-inch cyclotron at Lawrence Berkeley National Laboratory.

Whatever the future holds, the need for expertise in radiochemistry and nuclear chemistry is unlikely to wane. “Issues involving radioactivity will never go away,” says Roberts. “Even if we were to get rid of our weapons stockpile, we’d need to understand nuclear weapons and radioactivity for nuclear waste disposal, for example.” As they have demonstrated in the posttesting era, Livermore's radiochemistry researchers are ready to respond to new challenges as national needs change.

—Rose Hansen

Key Words: actinide, activation product, carrier-free isotope, Center for Accelerator Mass Spectrometry (CAMS), crown ether, extraction chromatography, fission yield, flerovium, Flerov Laboratory of Nuclear Reactions (FLNR), heavy elements, high-purity germanium radiation detector, isotope, livermorium, National Ignition Facility (NIF), nuclear chemistry, nuclear forensics, radioactive decay, radiochemistry, transactinide.

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Tiny Silicon Pillars Can Detect Giant Threats

A critical aspect of protecting our borders is detecting the entry of illicit special nuclear materials (uranium and plutonium isotopes). To address concerns about potential smuggling or misuse of nuclear materials, in particular in an improvised nuclear device, the Departments of Homeland Security and Energy are supporting efforts to develop a new generation of instruments for detecting the neutrons that fissile materials continuously emit. These efficient, compact, and modular detectors would, for example, be used to scan air and ship cargoes, investigate suspicious items, monitor spent nuclear fuel, help guard border crossings, and assist in ensuring compliance with arms control treaties.

A Lawrence Livermore research team led by electrical engineer Rebecca Nikolic has demonstrated a miniaturized, solid-state detection system that fulfills the need for a far more efficient and compact neutron detector than existing devices. The instrument, called a pillar detector, uses a detection element as thin as a credit card and manufactured primarily from silicon. Currently in advanced development, the device has demonstrated high efficiency without the many disadvantages of competing designs.

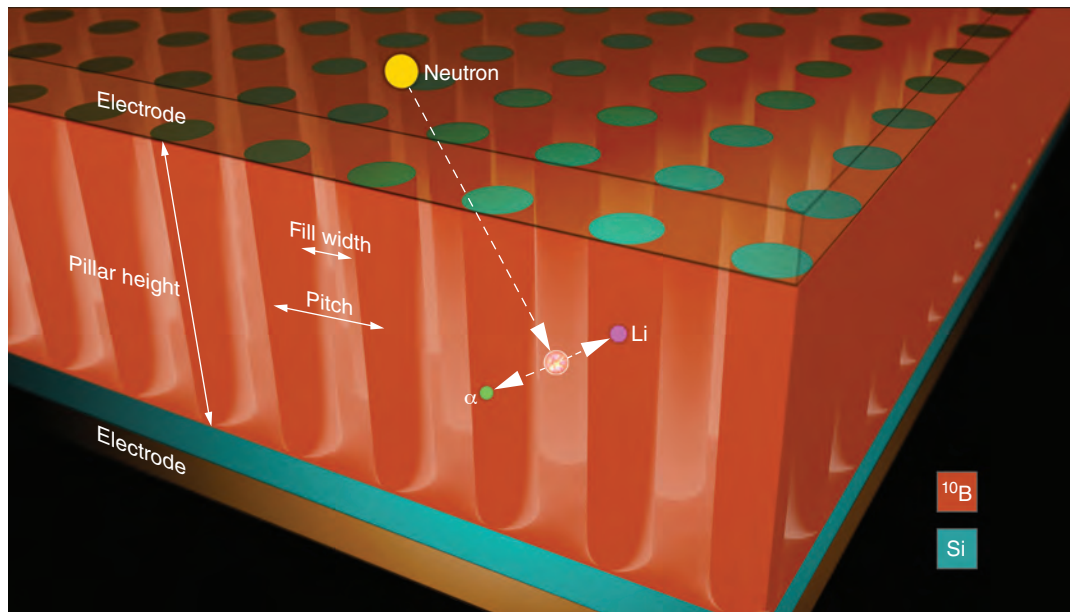
Of particular note, the pillar detector does not rely on increasingly rare helium-3, which current neutron detectors use to absorb thermal (low-energy) neutrons. Instead of helium-3, the

pillar detector uses a platform of etched silicon pillars interspersed with boron-10, an enriched boron isotope. Incoming neutrons from fissile materials strike the boron nuclei, yielding both alpha (helium nuclei) and lithium-7 particles. These particles travel a small distance before striking the semiconductor pillars to induce a measurable electrical current.

Multiyear Development Effort

Livermore's pillar detector research effort began in 2005 with funding from the Laboratory Directed Research and Development Program. The Department of Homeland Security's Domestic Nuclear Detection Office (DNDO) has supported the project since 2008, and the National Nuclear Security Administration's Office of Nonproliferation Research and Development has supported it since 2012. In addition to Nikolic, team members include Qinghui Shao, Lars Voss, Adam Conway, Catherine Reinhardt, Tim Graff, and Phillip Kerr. Their expertise ranges from nuclear physics to electrical engineering and materials science.

Working at Livermore's Center for Micro- and Nanotechnologies, the team applied advanced microfabrication techniques pioneered by the semiconductor industry to produce a breakthrough instrument that is both inexpensive to manufacture and low in maintenance.



In the Livermore-developed silicon pillar detector, incoming neutrons interact with the boron (^{10}B) “fill” between the pillars. When the resulting charged particles strike the silicon (Si), a signal is generated at the electrodes. Tall, narrow pillars increase the amount of ^{10}B available to detect neutrons. Shorter pillars improve the detector’s robustness, and wider pillars increase the detection of alpha (α) and lithium (Li) particles. The pillar pitch (distance between pillar center points), fill width, and height are optimized for maximum efficiency in a compact, low-voltage package.

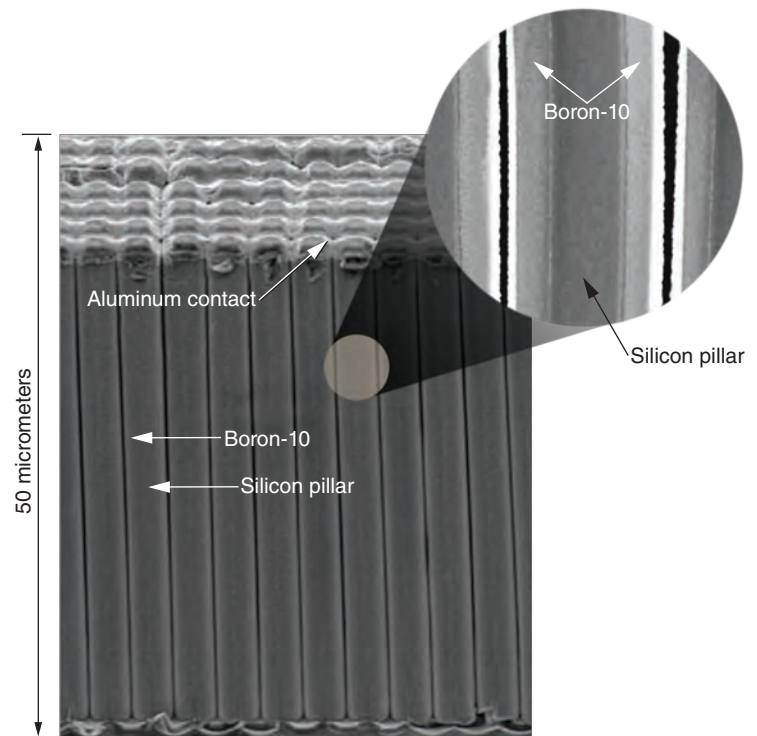
The project is one of many supporting the Laboratory’s nuclear nonproliferation mission.

Nikolic explains that because nuclear materials release both gamma rays and neutrons at different rates, they can be identified by their specific radiation signatures. For high-mass isotopes such as uranium and plutonium, being able to detect thermal neutrons is particularly important. “We need both thermal-neutron and gamma-ray detectors for positive identification of a special nuclear material,” says Nikolic. “However, detecting neutrons alone is a smoking gun. They indicate that special nuclear materials are likely to be present, and we can help to infer the identity of the fissile material from the neutron response.”

When neutrons are first emitted from a nucleus, they are very energetic but quickly slow down after colliding with surrounding materials, especially materials containing hydrogen. These thermal neutrons have an energy level (less than 1 electronvolt) and speed (about 2.2 kilometers per second) similar to that of room-temperature gas molecules and, thus, are more likely than energetic neutrons to be captured by a detector.

Current neutron detectors typically contain pressurized tubes filled with helium-3 gas to capture or absorb thermal neutrons. The tubes typically measure between 25 and 50 millimeters in diameter and up to 1 meter in length. Portable units must be robust, lightweight, efficient, and insensitive to vibrations and gamma rays. They also must be compact enough for handheld operation.

Comprising less than 5 percent the physical volume of a typical helium-3 detector, the pillar detector is ideal for handheld



The silicon pillar detector is only centimeters wide and requires less than 5 volts to operate. The detector is made with millions of etched silicon pillars, each measuring 50 micrometers high, and interspersed with boron-10. Both materials are readily available.

operation or other tasks where small size is critical. In addition, the pillar detector requires less than 5 volts to operate, while typical helium-3 detectors require a 1,000-volt power supply. “The pillar detector’s open-circuit voltage setting is similar to a solar cell,” says Nikolic. It achieves close to 50-percent efficiency in the laboratory and could be deployed in the same configuration in the field.

Nikolic notes that the search for a new generation of detectors has acquired some urgency because current supplies of helium-3 are declining rapidly. These supplies derive mainly from the dismantling of nuclear weapons, where the isotope accumulates as tritium decays. Instead of helium-3, the pillar detector’s primary materials are silicon (the mainstay of the electronics industry) and boron-10 (an abundant and easily separated isotope).

In searching for a substitute for helium-3, the team focused on boron-10, which has a large probability of reacting with neutrons. From 2005 to 2008, the team acquired a strong knowledge of the nuclear science of boron-10 as well as the multistep reactions that occur after a neutron strikes the boron, sending particles into the pillars that trigger an electronic signal.

Pillars Measure 50 Micrometers High

The key design feature of the new detector is millions of etched pillars measuring 50 micrometers high and spaced about 2 micrometers apart. The space between the pillars is filled

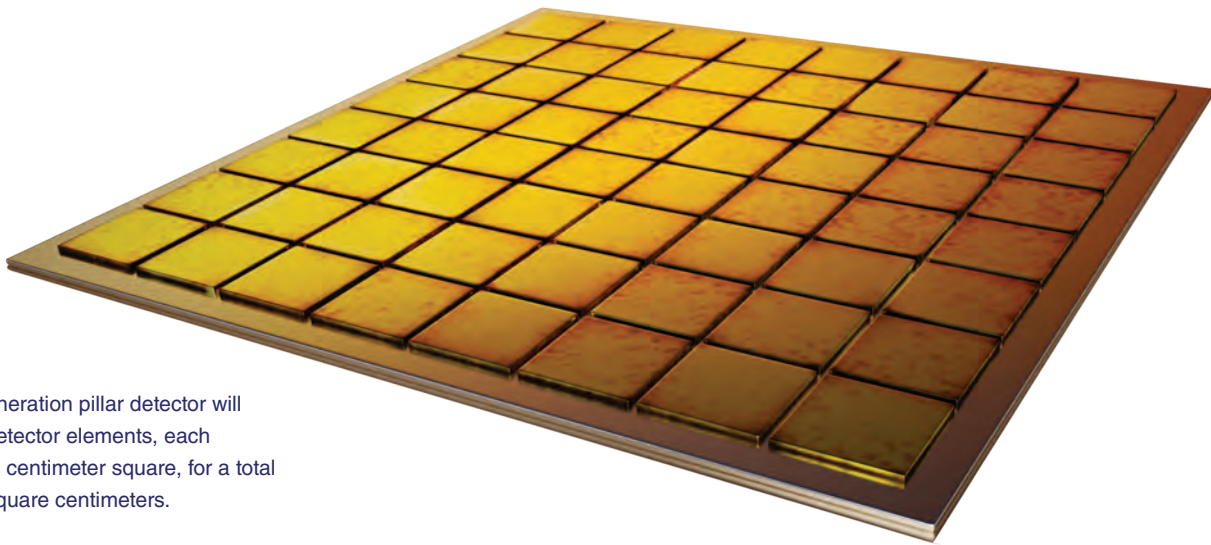
with boron-10 using a low-pressure chemical vapor deposition technique. The three-dimensional structure of the detector and the large amount of boron-10 maximize the capture of neutrons. The pillar height (or etch depth) can be increased to provide a thicker boron layer for capturing even more neutrons. “We can also adjust the spacing between the pillars so the alpha particles don’t have to travel far, which provides higher efficiency,” says Nikolic.

The detector relies not only on advanced manufacturing and assembly techniques but also on software to run the instrument, associated electronics (which are designed in collaboration with Lorenzo Fabris of Oak Ridge National Laboratory), and algorithms that analyze data in real time. The algorithms and hardware used to analyze the data from the pillar detector were originally developed for Livermore’s emergency response efforts by a team of scientists and engineers, including physicist Phillip Kerr. These technologies were incorporated into the Fission Meter, a neutron multiplicity detector based on long-standing helium-3 technology. The Fission Meter is now licensed to Ortec, Inc.

In 2013, a 16-channel (16-detector-element) design of the pillar detector successfully identified californium-252, a standard thermal-neutron source used to calibrate detectors. The detector measured the mass to 1-percent accuracy. The unit’s detection area of just 10 square centimeters completed the measurement in 18 hours, an eightfold improvement over a smaller prototype from a year prior. The team expects to achieve a less than 2-hour measurement time with a larger detector area by combining many individual detectors into an integrated, multichannel system. The next generation of the pillar detector features 64 detector elements,

In 2013, as part of a project for the Department of Homeland Security, Livermore researchers successfully demonstrated a pillar detector with 16 channels (16 detector elements), providing a detection area of 10 square centimeters.





The next-generation pillar detector will feature 64 detector elements, each measuring 1 centimeter square, for a total area of 64 square centimeters.

each 1 centimeter square, for an overall dimension of 8 by 8 centimeters (64 square centimeters).

The researchers also tested the pillar detector with cesium-137, a strong gamma-ray emitter. The neutron–gamma-ray rejection ratio was measured from 10^6 to 10^8 , which confirmed that the design does not confuse gamma rays with neutrons. Says physicist Steve Payne, “Importantly, the pillar detector is inherently insensitive to gamma rays and typically meets this requirement because its active region is very thin, only about 50 micrometers.”

Because the pillar detector is designed for strong neutron and gamma-ray radiation environments, the team conducted aging tests using Livermore’s Center for Accelerator Mass Spectrometry. Experiments with the center’s specialized beamline for studying the behavior of materials showed the pillar detector could withstand very high irradiation levels.

The pillar detector team has participated in three annual DNDO-sponsored neutron test campaigns. In these campaigns, helium-3 alternative technologies designed by national laboratories, industry, and academia are evaluated.

Applications from the Lab to the Field

Because the pillar detector design is modular, an instrument of virtually any size can be manufactured to meet the needs of different users. For example, a very small detector can be built for handheld applications, or many elements can be “tiled” to cover large areas. In stacking the detectors, one can achieve closer to 100-percent efficiency.

Potential applications range from the analytical laboratory to military, homeland security, and medical applications in the field. An immediate use would be for luggage and cargo container inspections at airports, seaports, and border crossings. Pillar detectors could find important roles in counterterrorism and arms control and safeguards.

Keeping Our Borders Secure

Nikolic expects the pillar detector to be licensed to a private firm. The team already holds several patents for the fabrication technology embodied in the device, and a few companies have expressed interest in the instrument. Nikolic anticipates additional interest from industry as the team demonstrates increasingly larger detector areas while decreasing the size of electronic signal processors.

The team is collaborating with researchers from Rensselaer Polytechnic Institute to refine the technique of depositing boron-10. In addition, the team is working on multiplexing (combining many electrical signals into one) the numerous small panels and miniaturizing the associated electronics to make possible a truly fieldable instrument.

Within a few years, Nikolic expects the detector to be performing double-duty in both radiation detection for counterterrorism and contamination control as it replaces far bulkier and less efficient detectors based on helium-3 in environments ranging from airports to particle accelerators. At the very least, the nation’s borders will be more secure, thanks to an exquisitely manufactured tiny device.

—Arnie Heller

Key Words: boron-10, Center for Accelerator Mass Spectrometry, gamma-ray detector, helium-3, pillar detector, silicon, special nuclear material, thermal neutron.

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Biosecurity

Gets a Boost with a Human-on-a-Chip

THE human body is a marvel—a complex, interconnected network of organs, tissues, and vessels controlled by chemical reactions and electrical impulses. So complicated is this system that many scientists and researchers dedicate their careers to better understanding how the body functions, adapts to outside stimuli, heals after disease or trauma, and reproduces to create new life. Advances in modern medicine, drug discovery in particular, rely on the continued study of the human body to develop treatments for health problems ranging from high cholesterol and chronic joint pain to the effects of chemical exposure. However, the drug development cycle, from initial research and formulation to clinical trials and commercialization, can take years. Even after

such lengthy study, about 90 percent of new drugs ultimately fail because they have unanticipated but serious side effects.

At Lawrence Livermore, a research team is working on an investigational platform that could speed development of medical countermeasures for biosecurity applications and improve the overall drug discovery process. The in vitro chip-based human investigational platform (iCHIP) combines primary human cells, tissue engineering, and microfluidics to reproduce the body's physiological response under an array of conditions.

"iCHIP is an integrated system for testing human response," says Satinderpall Pannu, who leads the iCHIP team of engineers, biologists, and chemists. "It includes electrical stimulation and

recording, nutrient and oxygen perfusion via fluidic delivery, optical analysis, and quantitative measurements of cell health and viability.”

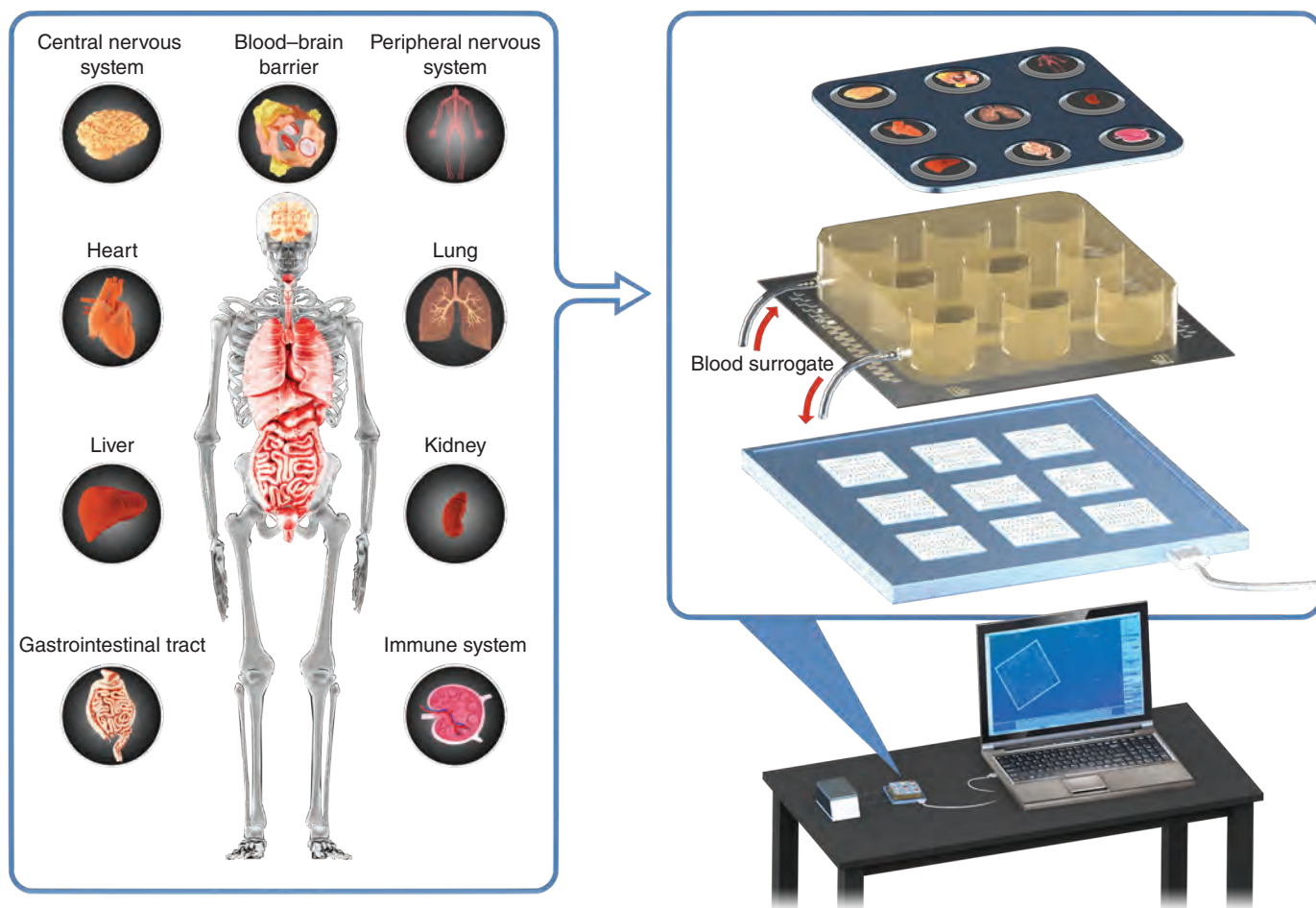
Thus far, the team has demonstrated the platform’s effectiveness on isolated human dorsal root ganglia (DRG) nerve cells, which form part of the peripheral nervous system. As envisioned, the complete iCHIP system will contain tissue from the stomach, liver, heart, kidney, brain, blood–brain barrier, immune system, and lungs, effectively creating what Pannu calls a “human-on-a-chip.” This technology has the potential to dramatically reduce the time needed to establish countermeasures against biological, chemical, and radiological agents and to bring new therapies to market.

From Retinas to Root Ganglia

According to Pannu, the current drug development process lacks an accurate and efficient experimental method for identifying whole-body effects of new pharmaceuticals. “Initial

tests are conducted on animals, and regulations require testing new compounds in two species before experiments move to humans,” says Pannu. “Animal testing also involves many ethical considerations, plus animal-based models do not accurately replicate the physiological response of humans.” For example, Vioxx, a medication for arthritis and other chronic pain, showed a protective cardiac effect in rats. However, five years after the Food and Drug Administration approved the medication, it was withdrawn from the market because it increased the risk of heart attack and stroke in humans. iCHIP is designed to replace the animal-based testing model in favor of one that is physiologically relevant to humans and takes into account a larger human population, providing a more realistic picture of a drug’s effectiveness and potential side effects in advance of clinical trials.

The iCHIP technology builds on Pannu’s previous work on a retinal prosthesis designed to function inside the eye. (See *S&TR*, June 2012, pp. 20–23; October/November 2009, pp. 14–15.) The



The integrated in vitro chip-based human investigational platform (iCHIP) will contain human tissue from each of the body’s major functional systems. Tissues are placed in tiny experimental wells on top of the microelectrode array.

artificial retina has a microelectrode array embedded in a flexible polymer, allowing the device to move naturally in the environment and conform to the live tissue around it. The microelectrode array in iCHIP is incorporated into a microfluidic device optimized to sustain cell health and is part of a suite of tests that can monitor cell activity. The microelectrodes stimulate and record tissue response; chemical sensors measure cellular function; and fluid exchange mechanisms deliver oxygen, nutrients, and test compounds to the tissue.

The iCHIP team, which is funded by Livermore's Laboratory Directed Research and Development Program, built its platform using DRG nerve cells—the sensory receptors of the body—to replicate the peripheral nervous system. DRGs are composed of sensitive neurons that are activated by pressure, temperature, and chemical stimuli. They thus serve as an excellent model for studying the neurotoxic effects of chemicals in the body. For the iCHIP research, the Livermore team obtains human DRG tissues through a partnership with AnaBios Corporation—a company with extensive expertise in human tissue-based assays. The DRG tissues are derived from organ donors, and unlike tissues grown from stem cells, they are completely mature. Their fully developed pathways provide a more reliable response to stimuli.

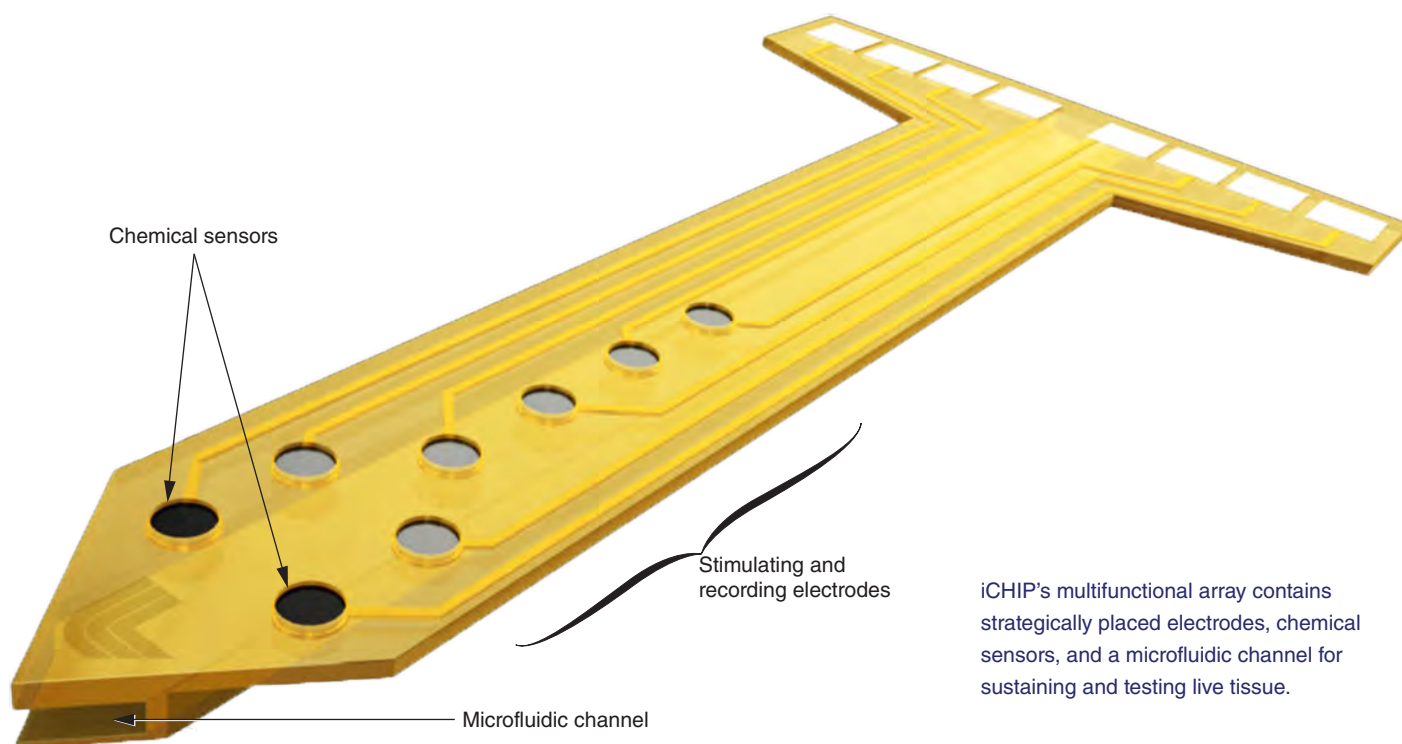
“One of our important achievements was increasing the viability of neurons on the platform from 2 days to more than 18 days,” says Livermore cellular biologist Kris Kulp, who leads the cell culture

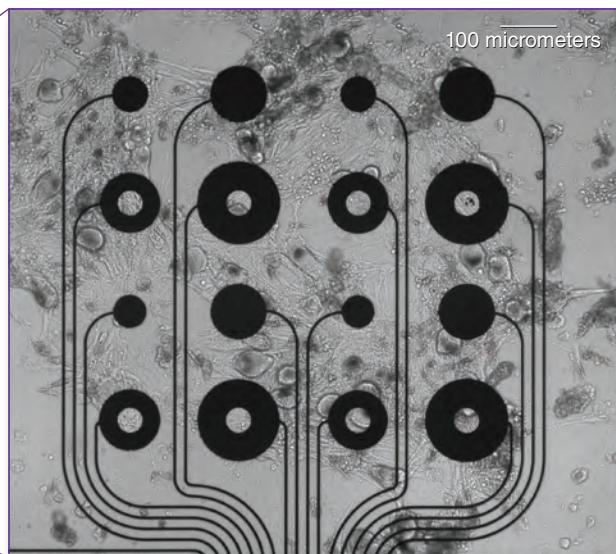
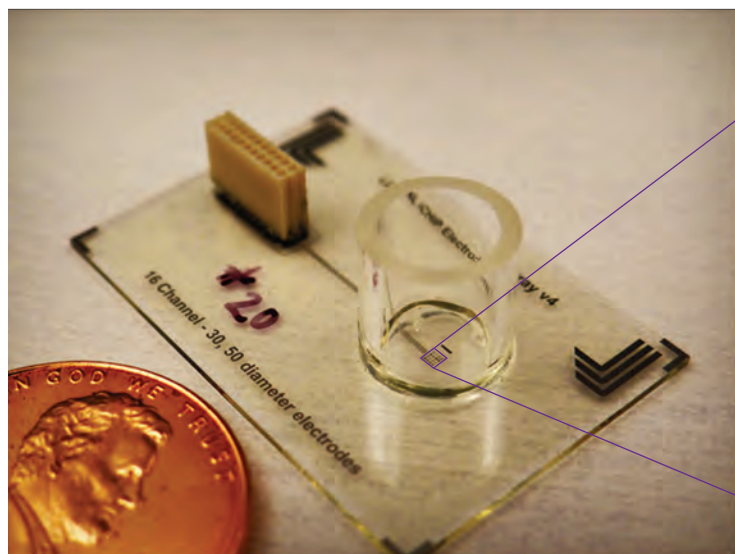
and assay development team. “We modified the surface chemistry of the platform to promote cell growth and adhesion, adjusted cell seeding density and the population of support cells, and tightly controlled media concentrations to extend cell function.”

The team is conducting experiments to interrogate cells for a prolonged period. The ability to monitor neuron response over time is a significant advance over traditional neurobiology assay methods, which rupture the cell membrane and kill the cells. Says Kulp, “By developing methods to repeatedly monitor cell response after chronic exposure to low levels of specific chemicals, we can model realistic exposure conditions and begin to develop countermeasures.”

Spicing It Up

In initial validation studies for iCHIP, the team exposed DRG cells to capsaicin—a component in chili peppers that is responsible for the burning sensation that occurs when specific nervous tissue receptors are activated. For the experiments, the human DRGs were plated into tiny experimental wells on top of the microelectrode array. “Traditional assays measure the response of only one cell at a time,” says lead platform engineer Elizabeth Wheeler. “With iCHIP, we can record the response of many kinds of neurons simultaneously, giving us a better understanding of a chemical's overall effect on the peripheral nervous system.”





Inside an experimental well, dorsal root ganglia cells are attached to a microelectrode array. Electrodes (black circles) stimulate and record tissue response.

Stimulating neurons triggers openings in the cell membrane, which control the movement of sodium, potassium, and calcium into and out of the cell. Ion movement across the cell membrane generates a voltage, or action potential, that can be measured. In the capsaicin experiment, the team recorded the resting and action potentials of cells exposed to low levels of the irritant for several minutes, noting changes in electrophysiology and calcium fluxes. “Our tests compared favorably to published results derived from traditional assay methods, suggesting that our platform will replicate the response of tissue *in vivo*,” says Kulp. “We will perform more validation assays to ensure that we can consistently achieve the right balance of neural and support cells and that the cells always respond the way we expect.”

For the next round of tests, the team will collaborate with the Laboratory’s Forensic Science Center (FSC) to measure tissue response to toxic agents and determine the mechanisms of toxicity. These experiments are the first step toward developing robust and effective medical countermeasures to known chemical agents. FSC is one of two U.S. laboratories that are internationally certified to identify chemical-warfare agents. Founded in 1991, the center supplies analytical expertise to counter chemical, nuclear, and biological terrorism and to verify compliance with international treaties.

Three-Dimensional Functionality

The iCHIP technology paves the way for bioengineers in the future to anticipate and understand new and emerging biosecurity threats and develop therapies that can be translated to humans

without adverse consequences. It also provides a foundation for designing human tissue-based assays to rapidly assess new drugs.

The project team is now focused on adding tissues from the central nervous system and the heart and on developing a three-dimensional vasculature scaffold, which will act as a “circulatory system” for the device. The scaffold will be built using additive manufacturing techniques and integrated with microsensors for more in-depth cell monitoring. “The scaffold will enable us to deliver reagents in a more realistic fashion,” says Pannu. “We will also be able to create a cell culture that contains more physiologically relevant cells and tissues to better answer questions about cellular processes, communication, and response to stimuli.”

In the future, the team wants to develop a method to either sustain multiple cell types in a single well or place them in electrically and chemically interconnected wells to diagnose intercellular reactions to stimuli. By building a human-on-a-chip, Pannu and his team are poised to revolutionize the way researchers analyze emerging biological and chemical threats and accelerate work on countermeasures to stop them.

—Caryn Meissner

Key Words: biological agent, biosecurity, drug discovery, *in vitro* chip-based human investigational platform (iCHIP), medical countermeasure, microelectrode array, microfluidic channel, pharmaceutical.

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Fuel Research Provides Insights into Basic Actinide Science

DEVELOPING new fuels for nuclear power plants is a dynamic business. Since the first nuclear power plant delivered electric power in the early 1950s, scientists have been searching for ways to improve the performance of fuels so that they burn more completely, thereby reducing radioactive waste and the risk that fuel could be used as a weapon. The March 11, 2011, disaster at the Fukushima Daiichi Nuclear Power Plant in Japan following the Tohoku earthquake and tsunami has increased concerns about the safety of nuclear energy. Nevertheless, it remains an integral part of the world's energy mix and accounts for 20 percent of the electricity generated in the U.S.

Despite 60 years of experience, improving fuel utilization, or burnup, has progressed slowly. The earliest reactors converted about 2 percent of their fuel into fission energy before the remaining material was identified as spent nuclear fuel. The

conversion rate for current nuclear power reactors is at most 4 to 5 percent. Researchers are developing a new class of fuels, known as ultrahigh burnup (UHBU) fuels, to increase the burnup rate at future reactors to 10 percent or perhaps even 20 percent.

Over the last three years, physicist Patrice Turchi in Livermore's Physical and Life Sciences Directorate has been refining the scientific methodology to accelerate the development of UHBU fission fuels. His team, which is funded by the Laboratory Directed Research and Development Program, includes experimentalists Luke Hsiung, Joseph McKeown, Michael Fluss, Mark Wall, Scott Tumey, and Thomas Brown and simulation experts Jean-Luc Fattebert, Alexander Landa, Vincenzo Lordi, and Per Söderlind. The project builds on Livermore's expertise in the actinide elements (those with atomic numbers 89 through 103) and focuses on uranium and plutonium. The team's goal is to understand how

the stability and microstructure of these radioactive elements evolve when they are mixed with other materials in both normal and irradiated environments. Such information will help nuclear engineers create mixtures with higher confidence in a new fuel's performance and longevity.

To date, the project team has worked in various Livermore facilities, synthesizing and characterizing minute quantities of radioactive specimens and exposing them to a process of accelerated aging to study their continued performance. The researchers also used the Laboratory's high-performance computers to simulate structural changes that occur in the actinide-based mixtures over time. In collaboration with Texas A&M University, they examined more than a dozen actinide-metallic alloys.

Transmission electron microscopy (TEM) images revealed features that researchers had not previously observed. For example, in uranium-zirconium alloys, two phases coexist at room temperature, and a structural and morphological change occurs after high-energy irradiation. The TEM images also helped clarify the structure of uranium-molybdenum alloys at room temperature.

A Close Look at Fuels

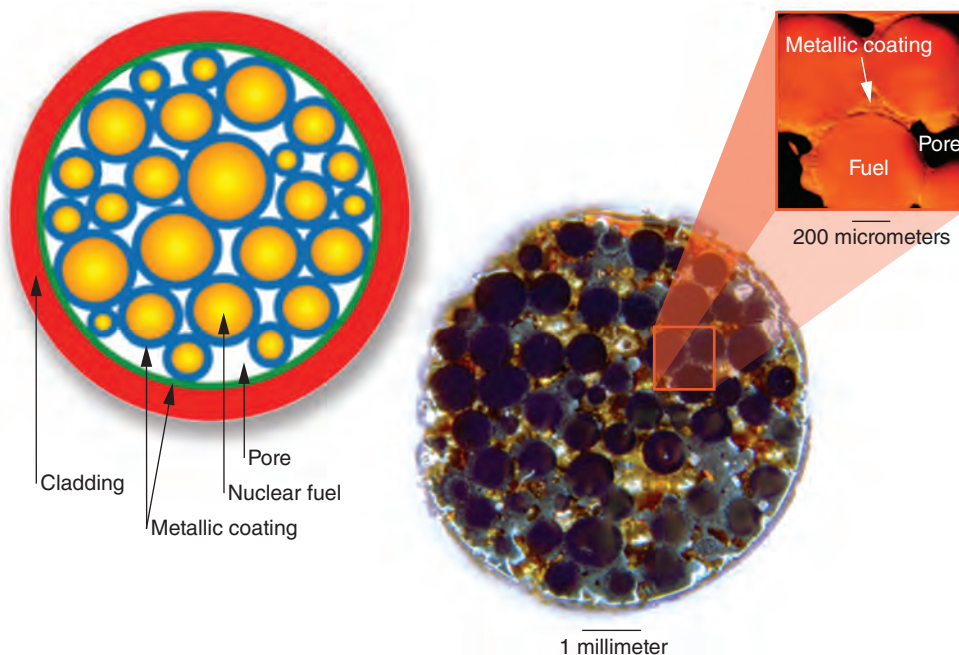
Top requirements for next-generation power reactors are optimizing nuclear fuel usage and minimizing waste, while making fuels more resistant to proliferation. In commercial reactors, as the uranium fuel burns, it constantly "breeds" plutonium, which is a major component of the spent nuclear fuel. These heavy elements are continuously fissioning, leaving behind fragments of many other elements, which are called fission products. The chemistry of the complex fuel mixture evolves with time, and the concentration

of fission products increases. Innovations in nuclear fuels focus on making optimal use of the uranium fuel as well as the plutonium and fission products, burning them up as much as possible to limit the radiotoxicity of the waste produced and reduce the potential for nuclear proliferation.

Inert matrix fuels show promise for improving burnup. These fuels have an actinide fuel kernel (a mixture of uranium with silicon or a metal such as molybdenum, niobium, or zirconium), an inert metallic matrix (such as a zirconium-based alloy), pores, and cladding materials for fuel containment. Experiments to date demonstrate that inert matrix fuels are highly resistant to irradiation and damage effects. The thermal conductivity of these fuels is also higher than it is in oxide-type fuels, which guarantees high heat transfer. Plus they protect against interactions with the cladding. In addition, says Turchi, "Inert matrix fuels represent an ideal system for conducting fundamental science research because each of the fuel's components can be studied separately."

Structure Predicts Performance

As part of the UHBU study, the Livermore researchers worked on developing computational techniques to predict the behavior of the new fuels. In the process, they broke new ground for nuclear materials science. Having a predictive capability is tremendously useful in guiding experiments, which are costly, and in further validating experimental results. Prediction demands accurate models whose results successfully stand up against empirical data. Part of the team's work, therefore, focused on quantum mechanical modeling—which examines the behavior of electrons—to compare simulated results with data from laboratory experiments.



A schematic (left) illustrates the components of an inert matrix fuel rod for nuclear power plants, including the outer cladding, an inert metallic coating, and nuclear fuel spheres. A mock inert matrix fuel (lower right) was fabricated for demonstration purposes using zirconium spheres as a fuel surrogate. The metallic coating is made of a zirconium-based alloy, and a stainless-steel cylinder provides the cladding. A higher magnification image (inset) shows the alloy coating the fuel spheres, which is important for high thermal conductivity, and porous regions, which accommodate fission gas during the reactor's operation.

An early study of zirconium-based actinide fuels, performed with colleagues at the Royal Institute of Technology in Stockholm, Sweden, showed excellent agreement between predicted energetics and experimental data. Later studies on a collection of molybdenum-based fuels, which examined how heat and magnetism influence the alloys, found similar agreement. Subsequent modeling focused on actinides—uranium, neptunium, plutonium, and americium—mixed with each other or combined with metals such as titanium, niobium, tantalum, and tungsten.

Results from the quantum mechanical studies provided input for assessing the mixtures with the CALPHAD (calculation of phase diagram) approach. CALPHAD efficiently describes the thermodynamic properties of all phases in a system. It also predicts the phase diagram of a fuel mixture, producing a graph of temperature versus composition to illustrate the stability of a material's various solid phases and its liquid and gas states.

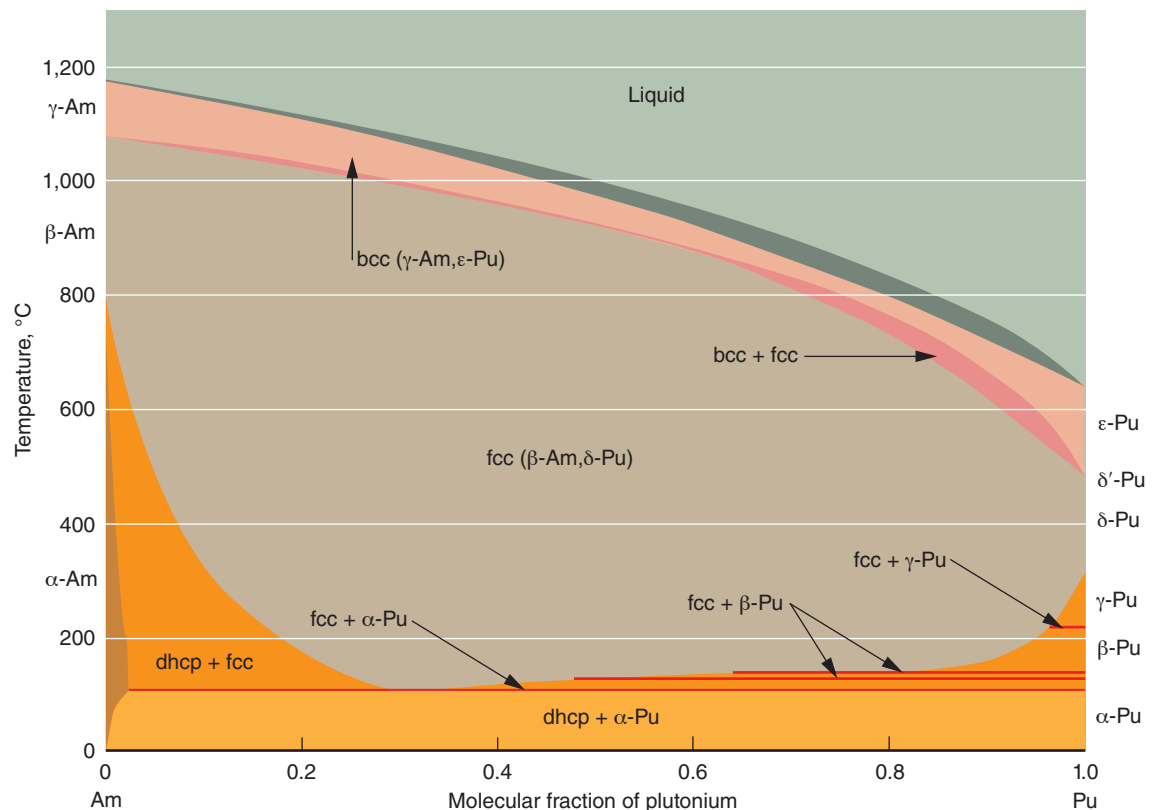
Phase diagrams are the basic road maps for designing alloys and understanding their behavior. They provide valuable information for material designers because structure controls a material's properties and hence its performance. The real

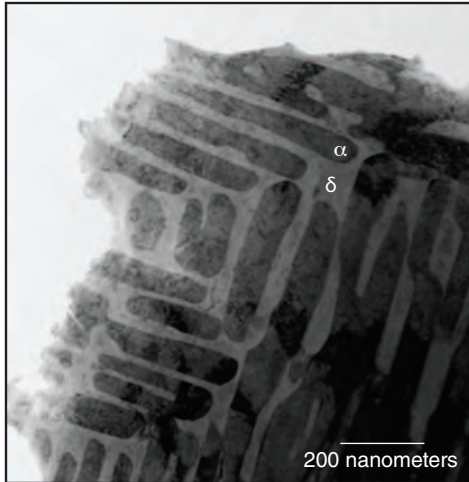
power of the Livermore methodology has been in predicting phase diagrams for multicomponent systems, including molybdenum–uranium–zirconium, molybdenum–plutonium–uranium, and even four-element combinations such as aluminum–molybdenum–silicon–uranium.

In experiments with researchers at Texas A&M University, the Livermore team characterized cast uranium–zirconium specimens using various experimental techniques such as x-ray diffraction, scanning electron microscopy, and TEM. Energy-dispersive spectrometry allowed the team to examine the local composition of nanometer-scale phases in each specimen. With these data, the team confirmed the coexistence of two equilibrium phases (alpha-phase uranium and delta-phase uranium–zirconium) at room temperature, representing the first experimental confirmation of this two-phase phenomenon in an as-cast, uranium-rich uranium–zirconium alloy.

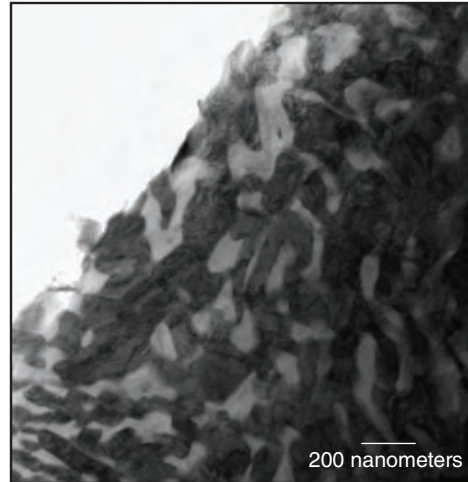
The experiments with cast alloys were primarily focused on assessing the thermal stability of the specimens prior to irradiation studies at Livermore's Center for Accelerator Mass Spectrometry (CAMS). A major challenge in studying the evolution of a nuclear fuel is the years required to irradiate a fuel in a reactor

This phase diagram, generated from energetics calculated with advanced quantum mechanical approaches and the CALPHAD methodology, provides a thermodynamic assessment of a binary alloy system made with americium (Am) and plutonium (Pu). Phase diagrams provide valuable information for designing alloys and understanding the new material's behavior. The Greek letters α , β , γ , δ , δ' , and ϵ indicate different material phases. The alloy's crystal structure may be double hexagonal close packed (dhcp), face-centered cubic (fcc), or body-centered cubic (bcc).





This micrograph of an as-cast uranium–zirconium alloy shows the coexistence of alpha-phase uranium (α ; dark contrast) and delta-phase uranium–zirconium (δ ; light contrast). The alloy has a lamellar, or platelike, microstructure with alternating lamellae of the two phases.



When the uranium–zirconium alloy was irradiated at the Center for Accelerator Mass Spectrometry, it lost its lamellar structure, as shown in this micrograph.

and measure its changes. At CAMS, researchers can emulate the consequences of accumulated fission damage by implanting heavy ions in samples at very high energies. Results comparable to years of irradiation are available in hours or days. In past experiments at CAMS, Livermore researchers simulated the aging of weapons-grade plutonium to determine whether helium bubbles that form over time would affect a weapon's performance. (See *S&TR*, September 2011, pp. 4–10.) This time, the purpose was to create defects in the alloy by implanting iron ions whose energy is similar to that of a fission fragment emitted by a nuclear power plant. Initial characterization displayed a loss of the material's well-aligned lamellar, or platelike, microstructure. Says Turchi, "This change may indicate that the alloy loses its dimensional stability, which could affect the fuel's performance."

Continued Collaborations

In addition to producing a host of actinide science advances, the UHBU fuel research has led to a number of spin-offs. For example, Livermore metallurgist Adam Schwartz is leading one of the four focus areas for the Department of Energy's Critical Materials Strategy Initiative, investigating substitutes for rare-earth elements and other critical materials used in clean-energy technologies. The tools developed to study actinides are being applied to examine the rare-earth elements, which occupy the row above the actinides in the periodic table and share many features with that group.

In 2013, Turchi was one of two Americans asked to participate in developing an international database on the thermodynamics of advanced fuels for the Organisation for Economic Cooperation and Development–Nuclear Energy Agency. The goal of the three-year project is to develop a flexible computational tool using CALPHAD to perform thermodynamic calculations on different types of fuels,

fission products, and structural materials such as steel and concrete for the next generation of nuclear reactors.

As part of a collaboration between the National Nuclear Security Administration and CEA, the French center for atomic energy, the first CEA fellow to work at the Laboratory arrived in October 2013 for a two-year assignment. Aurélien Perron is modeling the thermodynamics of plutonium alloys that are relevant to stockpile stewardship and advanced nuclear fuels, using this information to carry out kinetic and microstructural studies.

Turchi adds that, although his colleagues never intended to explore conventional nuclear fuels, their findings may prove useful to improving the use of those fuels. The Department of Energy's Global Threat Reduction Initiative aims to replace highly enriched uranium (HEU) in research reactors around the world with low-enriched uranium (LEU). HEU is much easier to weaponize than LEU and thus is considered a proliferation threat. The team studied a uranium–molybdenum alloy that is a leading LEU candidate, and those results could prove useful to engineers and policy makers who will evaluate the feasibility of converting reactors to an unproven fuel. Transitioning to LEU fuels may expand the number of countries that can host reactors for scientific research and produce isotopes for medical and other peaceful applications.

—Katie Walter

Key Words: actinide–metallic alloy, CALPHAD (calculation of phase diagram) approach, Center for Accelerator Mass Spectroscopy (CAMS), rare-earth element, transmission electron microscopy (TEM), ultrahigh burnup (UHBU) fission fuel.

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Patents

Virtual Gap Dielectric Wall Accelerator

George James Caporaso, Yu-Jiuan Chen, Scott Nelson, Jim Sullivan, Steven A. Hawkins

U.S. Patent 8,575,868 B2

November 5, 2013

A virtual, moving accelerating gap is formed along an insulating tube in a dielectric wall accelerator by locally controlling the tube's conductivity. Localized voltage concentration is thus achieved by sequential activation of a variable resistive tube or stalk down the axis of an inductive voltage adder, producing a virtual traveling wave along the tube. The tube's conductivity can be controlled at a desired location, which can be moved at a desired rate, by light illumination, photoconductive switches, or other means. As a result, an impressed voltage along the tube appears predominantly over a local region, the virtual gap. The effective gain of the accelerator can be made very large by making tube length large compared to the virtual gap length.

Laser Fusion Neutron Source Employing Compression with Short Pulse Lasers

Joseph A. Sefcik, Scott C. Wilks

U.S. Patent 8,576,971 B2

November 5, 2013

This method for achieving fusion includes a laser source that generates a laser beam and a capsule filled with deuterium–tritium (DT) gas embedded in a target. When the laser beam is directed at the target, it helps create an electron beam within the target. The electron beam heats the capsule, the DT gas, and the area surrounding the capsule. At a certain point, equilibrium is reached, and the capsule implodes, generating enough pressure to ignite the DT gas and fuse the gas nuclei.

Awards

Ken Moody, the Laboratory's chief scientist for radiochemistry, was named a fellow of the **American Association for the Advancement of Science (AAAS)**, one of 388 AAAS members to be elected this year. Moody joined the Laboratory in 1985 and has been a critical member of the team that discovered six new elements—113 through 118. (See the box on p. 10.) Trained under Glenn Seaborg, Moody has dedicated his career to the scientific advancement of radio- and nuclear chemistry. In addition, Moody is one of the creators of the discipline of nuclear forensics, and applications of radiochemistry to national security and law-enforcement problems.

The **American Physical Society (APS)** named four scientists in the Laboratory's Physical and Life Sciences Directorate as **2013 fellows**.

Charles Cerjan was cited by the Division of Atomic, Molecular, and Optical Physics for "seminal contributions to time-dependent Schrödinger equation propagation algorithms and their application to particle scattering and intense field dynamics, the development of laser-produced plasma sources for advanced lithography and the investigation of the basic mechanism of magnetic multilayer material response and its application to magnetic storage devices." Since joining the Laboratory in 1983, Cerjan has worked on soft-x-ray lithographic sources, magnetic materials for hard disk drive applications, intense laser field effects, and nuclear diagnostic analysis for the National Ignition Facility (NIF).

Marilyn Schneider was recognized for "outstanding contributions to X-ray measurements in laser-produced plasmas" by the Topical Group on Instrument and Measurement Science. Schneider joined the Laboratory in 1986 and is now group leader for Radiative Properties and functional group leader for Radiation Physics and Spectroscopic Diagnostics in NIF.

Eric Schwegler was honored by the Division of Computational Physics for "important contributions to the development of linear scaling electronic structure theory, and the use of first-principles methods to examine the properties of aqueous solutions, nanomaterials and matter under extreme conditions." Schwegler came to Livermore as a postdoctoral researcher in 1998. In 2005, he became the group leader for Quantum Simulations.

Ian Thompson was cited by the Division of Nuclear Physics for "the development and application of all-order treatments of nuclear-cluster dynamics in peripheral reactions; leading to a new understanding of halo nuclei within a few-body framework." Thompson joined the Laboratory in 2006, after serving as professor of physics at the University of Surrey, England. He also is a fellow of the United Kingdom's Institute of Physics.

The four researchers join Livermore scientists John Moody and Pravesh Patel, who were recognized earlier this year. (See *S&TR*, January/February 2014, p. 20.) In the past 30 years, nearly 100 Laboratory employees have been elected APS fellows. Election to APS fellowship is limited to no more than one half of 1 percent of the society's membership for a given year.

Radiochemistry Renaissance

Over the past few years, Lawrence Livermore has experienced a surge of innovation in radiochemistry—the science of generating, identifying, isolating, and characterizing the radioactive isotopes of chemical elements. For example, Livermore researchers have developed a technique for producing double-sided target foils and built a target surface mapping tool, which increased the productivity of stockpile stewardship experiments. Radiochemists are also using the Laboratory's particle accelerator to measure nuclear reaction products for reactor safety and nuclear forensics research. Another Livermore team is making synthetic nuclear debris samples with ingredients that more closely match debris from a real explosion for use in isotope identification exercises. Researchers are also refining the chemicals and protocols required to perform wet-chemistry experiments on transactinides, short-lived heavy elements such as flerovium and livermorium. A collaboration with two universities is developing a portable, automated system that can quickly capture, prepare, and chemically test radioisotopes.

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Predicting Wind Power with Greater Accuracy



Researchers are combining fieldwork, advanced simulations, and statistical analyses to help wind farm and electric grid operators.

Also in April/May

- *Livermore researchers combine first-principles calculations with high-performance computing to model low-energy nuclear reactions at a fundamental level.*
- *Understanding why a common bacterium can thrive in uranium-contaminated settings may boost efforts to develop new toxic-waste remediation strategies.*
- *A clever computational method speeds up the modeling of chemical systems a thousandfold over traditional methods.*

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